Progress Report & Renewal Proposal

ONR, 1994 Renewal of Contract #N00014-89-J-1227

J. L. Hall, August 1994 University of Colorado

1. Research Description. This ONR funding provides essential support for a vigorous ongoing research effort involving approximately 11 people, including 4 postdoctoral researchers, 4 graduate students, and two visiting faculty members. The focus of the work is the development of precision laser techniques and their application to physical problems of outstanding scientific interest, including laser frequency stabilization at/below the Hertz level, cold atomic "fountains", optical frequency standards, ultra-resolution spectroscopy, measurement of optical frequencies, and advanced tests of physical principles. The main thrusts in the near future are to complete our frequency measurements of the stabilized Nd laser, bring the stable lasers together with the slow atoms, and to demonstrate a new and precise spectroscopic technique based on intracavity molecular dispersion in overtone bands.

2. **Scientific Problem**. We are interested in the coherent interaction of atomic and radiation systems, with an eye to increasing the coherent interaction time via laser/atom beam slowing techniques, Ramsey fringe techniques, and the still-being-refined atom trap/atom fountain techniques. Application directions include optical and microwave atomic frequency standards, atomic interferometry, and a new concept for testing the electrical charge neutrality of atoms. A second main interest focusses on modifying the statistical character of radiation fields, including generation of squeezed light and correlated photon beams using nonlinear optics, generation of laser fields of reduced relative phase diffusion using special lasers designed to exhibit correlated spontaneous emission, and generation of apparently-random fields by electro-optic modulation with a deterministic "noise" source. Applications include ultrasensitive detection of weak absorption, possible enhancement of sensitivity of laser gyro and gravitational wave sensors, and enhanced broadband detection of multiplexed absorption spectra.

Of course, at any particular time these generalities are manifested as specific interests and experimental areas, with the present activities elaborated more fully below. For example, an exciting new possibility under active consideration is the measurement of the 4-vertex QED process which could be called "light by light scattering," but which will be measureable as a birefringence of the vacuum, induced by a string of 6 SSC magnets (6 Tesla $\rightarrow \Delta n/n = 2 \times 10^{-22}$). This QED measurement will be based on the enabling technology of accurate locking to a high-finesse cavity, developed for our successful experiments with cavity-stabilized He-Ne lasers. A collaboration with other academic and national labs people is projected.

3. Scientific & Technical Approach. Using the developing tools of ultrastable laser, optical frequency measurement and microKelvin atomic beam technologies, we are exploring the prospects for advanced physical measurements. One of the most interesting challenges at present is to bring together the developing bits of ultrastable laser and microKelvin atomic beam technologies to build an optical frequency standard of unprecedented performance. We expect an optical Ramsey resonance lineshape of about 10 Hertz width at a center frequency of about 10^{15} Hertz, and the large signal to shotnoise ratio characteristic of utilizing some million atoms, namely ~1000:1. The optimum servo control of the laser using this signal could evidently produce a laser linewidth in the domain of a dozen milli-Hertz! What is particularly attractive about this system is the expectation of extremely high accuracy as well, because of the absence of incalculable velocity-dependent shifts, combined with the absence of collisional effects. Thus the inaccuracy of such an optical standard must surely be in the sub-Hertz domain. Magneto-Optic trapping techniques in a sealed cell, coupled with our advances in stabilizing diode lasers can lead to more simple and portable realizations of these techniques in the future.

A large number of interesting scientific problems will be available with such a source. We foresee atom interferometers with frequency readout of infinitesimal Sagnac-induced or gravitational-gradient-induced phase-shifts, atomic velocity measurements at the level of a few micons/sec which lead in turn to a drastically improved limit for the charge neutrality of atoms and, potentially, a useful measurement of the gravitational response of individual atoms. The wave-mechanical nature of the atoms themselves also gives rise to interesting and potentially useful quantum interference effects. A test of T symmetry via setting a better limit on an atomic electric dipole moment may be feasible in the future. Measurement of the frequency of this atomic fountain reference against the microwave frequency standard opens the door to exciting further tests of our fundamental physical postulates, such as the invariance of atomic frequency ratios in time or against changes of gravitational potentials and/or fields.

Our successful experiments with cavity-stabilized He-Ne lasers provide one of the main enabling technologies for this precision work, namely optical sources locked with sub-Hertz precision. Frequency unpredictability is in the few Hertz domain even after a few minutes. Secondly, for precision atomic spectroscopy, a useful sample of low-speed atoms can be produced by the laser-cooling, using techniques which we and others have been developing intensively. The present method of choice is to capture slow atoms in a gas cell in a zone of inhomogeneous magnetic field -- the so-called "sealed-off Magneto-Optic Trap." This process results in an sodium atomic sample with a diameter ~ 1 mm at a kinetic temperature below 100 milliKelvin. The magnet supply current for the inhomogeneous field is then switched off, and the molasses laser intensity lowered to produce a much lower atom temperature, below 40 μK kinetic temperature.

As a concrete spectroscopic application, in the thesis project of Mr C. Oates we are then making a further optical pumping and Raman velocity-selective transition to prepare in F" = 2 a velocity subgroup of atoms with only a few cm/s velocity dispersion. These sodium atoms are then weakly excited with D_2 light on the cycling transition. Measurement of the limiting linewidth at low intensity gives directly the frequency equivalent of the atomic lifetime, with an inaccuracy which is expected to be ~0.2%. A present objective is to learn how to deal with the recoil shifts and lineshape issues with sufficient accuracy. It is interesting that in cw frequency-based spectroscopy, known experimental defects have the consequence of broadening the measured linewidth, whereas the situation is that our measured linewidth is too narrow according to the best many-body calculations[1]. Figure 1a shows a representative lineshape and fit, while Fig 1b shows theoretical predictions and other measurements.

These experiments with 1D cooling are also a directly useful preparation for our atomic fountain atomic clock experiment. There, in the final stages of cooling, the two vertical beam frequencies are changed, to provide a vertically-upward velocity reference in the molasses, thus ejecting the atomic 'pellet-like' sample upward for the atomic fountain. We are still agonizing over the decision of whether to pursue the fountain measurements with Ca, Ag, or Sr. In the former case things are basically in order and we may begin, but unfortunately a "forbidden" magnetic spin flip, electric dipole transition limits the lifetime of the target $^1D_2$ state, so that ~70 Hz is the narrowest linewidth that can be expected. In Ag the metastable state lifetime will offer a sub-Hertz future linewidth, but unfortunately the cooling radiation must be at 338 nm. At present this can best be obtained by harmonic generation of a tunable dye laser. We have recently experimented with vastly-improved $LiIO_3$ crystals which become available for efficient uv generation in an external ring resonator. Sum frequency generation with a powerful Ti:Sapphire laser might be interesting also. One concern with the two-photon approach needed for both these atoms is the intrinsic ac Stark shift, which is basically of the same magnitude as the two-photon Rabi frequency. The Ramsey separated oscillating fields approach helps this situation but may still leave an uncomfortable sensitivity.

So at present, we are tentatively designing around the use of Strontium as the atomic system, with cooling on the 460.9 nm E_1 resonance line (reached by frequency-doubling Ti:Sapphire with a-axis $KNbO_3$), and using the ^1S-$^3P_2$ M_2 transition at 671.2 nm for the clock. This decision means that we should be free of any important "light shifts" but we must be able to do velocity selection by saturation spectroscopy on this M_2 transition. Estimates of the needed power are comfortable (5 mW into a 30x build-up cavity) because of the long interaction time. As reasonable diode lasers are available now at both the needed wavelengths, it appears possible that in the future these radiations could be obtained from diode laser sources.

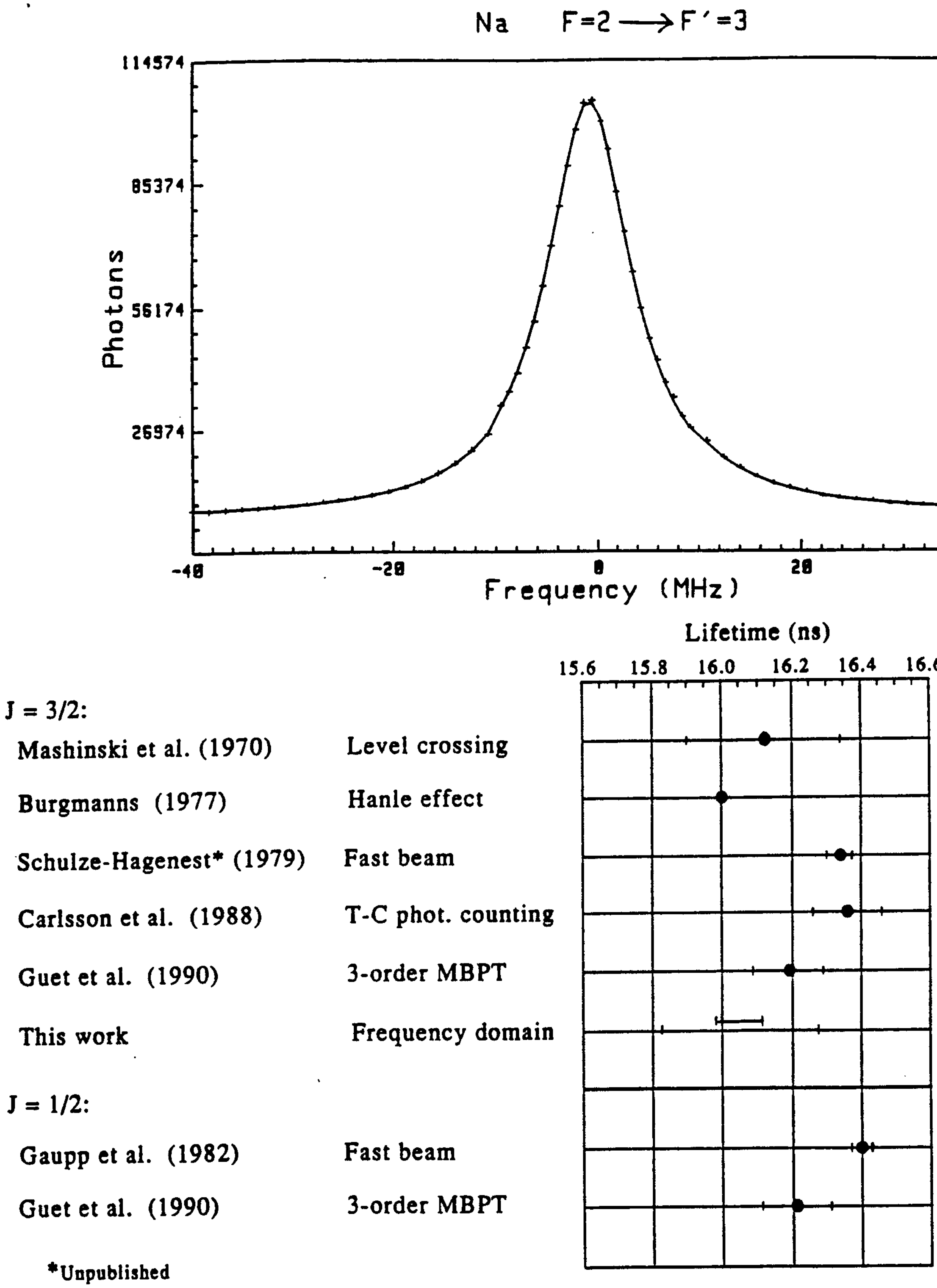

Figure 1. Lifetime comparisons for Sodium 3P. Top: Measured profile and least-squares fit. Lower: lifetime predicted by many-body calculations, along with various experimental results.

Unfortunately the investment in all the alignment and turning mirrors required is above $5000 so we will not start the real fountain experiment until the atomic species -- and hence effective wavelength -- is decided by the outcome of the doubling and mixing experiments, and the presently-undertaken stabilization experiment with 670 nm laser diodes.

As detailed below, we continue working with another system, based on the frequency-doubled Nd laser and Iodine absorption, which offers somewhat lower performance with vastly lower complexity. This is probably the best of the "good enough" systems which has been demonstrated, and we have shown that a frequency reproducibility ~300 Hz is readily obtained. A low-cost future prospect is a 633 nm laser diode working with the Ne* atomic $1S_5$-$2P_8$ transition, which remarkably is only 468 GHz from the usual HeNe standard. (We have already measured beats above 620 GHz.)

A new postdoc, Dr. J. Noh from Mandel's group in Rochester is working on an OPO-based scheme to generate "twin" photons for use in an interesting interferometry experiment. This idea is to follow up on a recent calculation by Holland and Burnett[2] in which amplitude-correlated beams are to be used in a Mach-Zehnder interferometer. The physical prediction is that phase can be better determined under these conditions than would be the case for coherent light. In the coherent case one expects the $\delta\Phi * \delta n = 1$ uncertainty relation to give $\delta\Phi * \sqrt{n} = 1$, whereas with amplitude-correlated light they predict $\delta\Phi * n = 1$. For a usual beam with 10^{16} quanta, this represents a fantastic change of the projected sensitivity. Another idea that will be tried is to use a frequency-doubling resonator with two tight-focus waists and two doubling crystals. Mlynek and his associates have recently demonstrated "bright squeezing" from a single crystal device[3]. We project that our two-crystal scheme will give us a pair of correlated beams, or indeed as many independent-but-quantum-correlated beams as we like with more crystals! This idea urgently must be tested.

4a. Progress:

Powerful diode-laser-pumped Nd:YAG lasers have become available commercially with exceptionally good frequency stability[4] and, after frequency-doubling, they can be tuned to at least 6 strong absorption lines in molecular Iodine. Two independent lasers systems have been stabilized to the 532 nm Iodine resonances using our (patented) "modulation transfer" spectroscopic technique[5]. Very strong resonances of nearly ideal symmetry are obtained -- see Fig. 2a. The Allan variance of the beat between two dissimilar systems is shown in Fig 2b and represents a stability unprecedented for any visible laser: σ_y ~60 Hz (1×10^{-13}) at 0.1 sec , improving below 4×10^{-14} for T> 20 sec. The

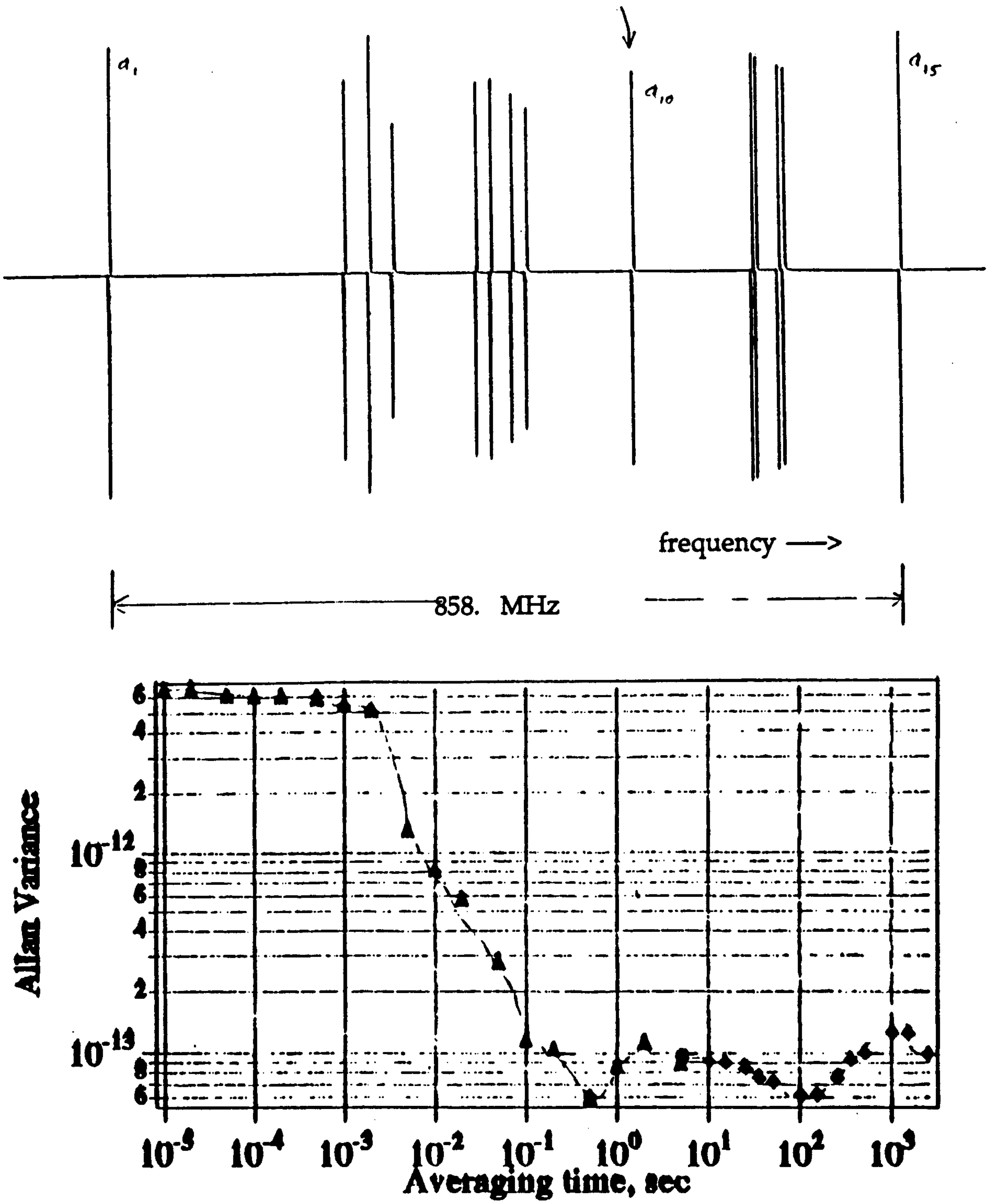

Figure 2. (a) Hyperfine spectrum in R(56) 32–0 line of I_2 at 532 nm, excited by frequency-doubled Nd:YAG laser, using modulation-transfer spectroscopy. (b) Allan variance of beat between two systems locked on a_{10} hfs component.

reproducibility at present is ~300 Hz, about 1 $\times 10^{-3}$ units of the operating linewidth. These first results with the doubled-Nd 532 nm green are some 40-fold better than those obtained with the 633 nm Iodine-stabilized HeNe laser, and this is after 24 years development work on the 633 nm system in countless national standards labs!

We have built a reliable apparatus using nonlinear crystals $LiNbO_3$ and $LiIO_3$, respectively, to double 1.06 μm, and to add the resulting green and ir frequencies, obtaining at present 1/4 mWatt of the 355 nm output. This will be heterodyned with the doubled output of a Ti:Sapph laser at 710 nm, which can be locked to various Bromine lines in this region. However, a delicious "target of opportunity" has recently come into view based on the excellent performance of this Nd/I_2 locking system, coupled with a new measurement of ±5 kHz accuracy reported recently by Nez, Clairon, Biraben, Felder et. al[6]. of the frequency of the 778 nm two-photon lines in atomic Rb. Remarkably, it turns out that these frequencies satisfy the equation:

$$2 * f(633\text{ nm}) = f(532\text{ nm}) + f(778\text{ nm}), \text{ to within 1200 GHz!}$$

One could view this as an experimental test of the final stage of T. Hänsch's "divide and conquer" scheme for establishing optical frequency differences spanning from the visible to the microwave range. This method leads here to an effective avenue for accurately determining the frequency of the Nd-green Iodine reference lines. However, for simplicity, we have made our measurements first relative to the Rb D_2 line at 780 nm, since the beat frequency is then "only" 263 GHz. The optical frequency of this transition is known to ±60 kHz from precise wavelength measurements[7] at NPL.

Our setup is illustrated in Figure 3. For the nonlinear optical system, we use doubling of the 633 nm laser with a temperature-tuned phase-matched RDP crystal in a "build-up" resonator, and do frequency-summing of the ir and green single-pass in an angle-tuned sample of the same material. The observed heterodyne beat S/N >20 dB in a 300 kHz bandwidth is more than adequate for the phase-tracking circuits we fabricated. The 263 GHz interval between two Ti:Sapph lasers at 780 nm has been measured with a "conventional" Schottky diode (fabricated for sub-mm radio astronomy), driven strongly at 43 GHz by a phase-locked klystron microwave source. The resulting 1 GHz beat has sufficient S/N for direct counting, but for safety we use a tracking oscillator here as well. The measured frequency of the Iodine molecular reference line is found to be

$$f(a_{10}\text{ component}) = 563\,260\,223.480\text{ MHz} \pm 70\text{ kHz}. \quad (\text{ R(56) 32-0 })$$

Basically all the uncertainty added by our frequency measurements arises from experimental insecurity in defining the "center" of the Rb transition. This line is 6 MHz wide and exhibits some interesting distortion/shift effects at

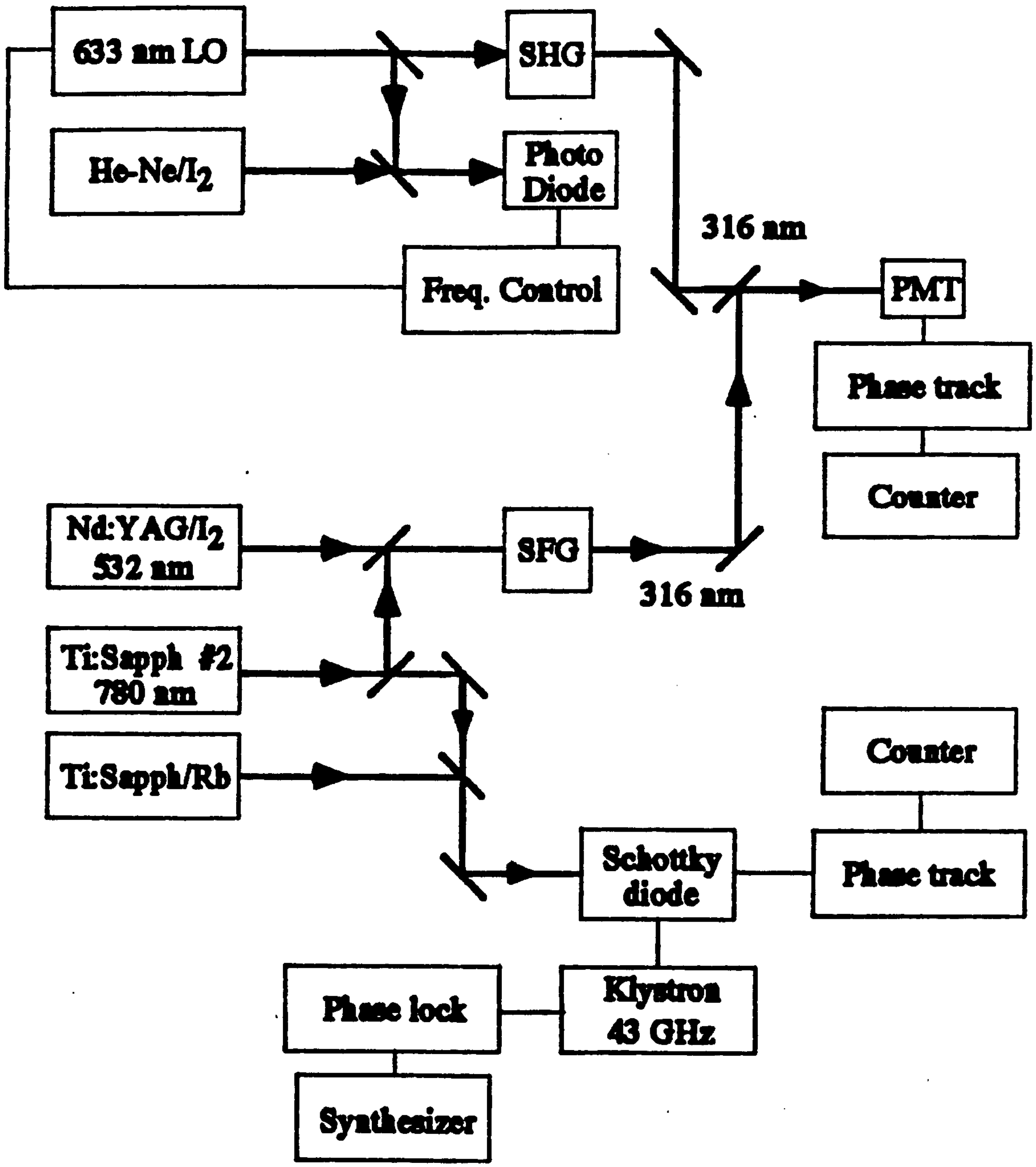

Figure 3. Experimental schematic for frequency measurement.

even modest power levels due to momentum transfer in a nearly-closed 2-level system. The "cross-over" lines are less affected. We have characterized this shift quantitatively based on the zero-third-derivative locking system employed. A more detailed study is planned to expand the work[8] of Grimm and Mlynek: see below. When we can measure the 1.2 THz beat associated with the narrow Rb two-photon line, this uncertainty will basically disappear, leading to frequency uncertainty in the range ~10 kHz.

For this we are pursuing an optical comb generator following the work of Ohtsu and Kourogi[9] who put a microwave phase-modulator in an optical cavity. We are now generating good sidebands, limited presently by temperature-induced dispersive effects. A Peltier-cooled/stabilized microwave structure is almost ready to try out. The Japanese workers have already seen modulation sidebands beyond 4 THz! Reduction of the phase noise of the best present rf source will be required for this approach to reach these very high frequencies with useful carrier/pedestal power ratios. We have the enthusiastic cooperation of F. L. Walls at NIST in this endeavour.

An interesting projected application of our precision frequency synthesis capability is to measure the frequency splitting of a two-photon line in He (2^3S–4^3D_1) which is only 224.2 GHz from the HeNe standard. Of longer term and more fundamental interest will be measurement of the energy level shifts over a series of Rydberg states to see the role of retardation in the Coloumb interaction. It is remarkable that contemporary many-body calculations[10] for He are adequate for such precision tests[11]. We are also considering trapped Li, for which the Rydberg levels could be reached with doubled Ti:Sapphire.

In the course of comparing the stabilized output of this Nd:YAG laser with our precise reference cavities –which are located two rooms away in our "Quiet House" – we discovered that 25 m of ordinary sheathed mono-mode polarization-maintaining fiber[12] can introduce a tremendous amount of noise onto an otherwise phase-stable light beam: more than 1 kHz of spectral broadening was written onto the light by phase-noise variations of the fiber's transmission phase. The perturbations include acoustic noise, micro-bending induced by vibration, and micro-thermal variations. With a frequency-offset coding at the remote end on a weak returned beam, by heterodyne at the source end we obtain two units of the fiber's insertion phase. This is used with a phase-locked VCO and digital divide-by-two to form a servo system using an additional AOM, accurately "pre-cancelling" the phase noise before the beam enters the fiber. In this way it was possible to reduce the fiber's output bandwidth from *kilo*Hz to the sub-*milli*Hertz level. Patent disclosures have been submitted to both CU and NIST on this work. The paper should appear momentarily in Optics Letters. We are attempting to organize with US West to provide a fiber link between NIST and JILA with which we will be

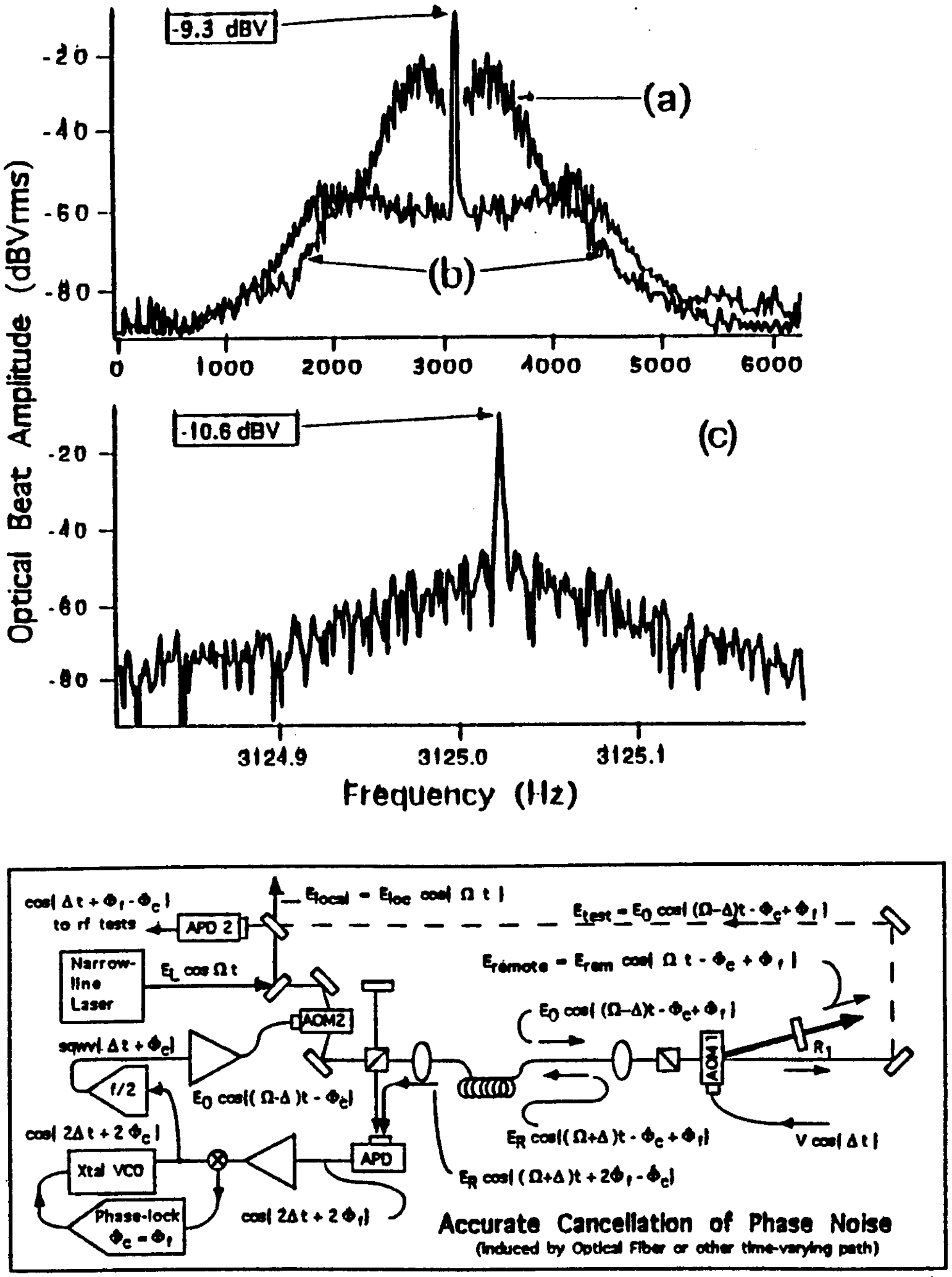

Figure 4. Lower part shows apparatus for accurate cancellation of fiber-induced phase noise. The one-way fiber noise is Φ_f, becoming 2 Φ_f on double pass. Phase lock regenerates beat signal to give signal for driving AOM-2 with accurately-cancelling signal, since the correction phase Φ_c closely equals Φ_f.

Upper part shows optical field spectrum after 25 m of fiber. (a). Original "delta-function" signal arrives with 1 kHz width. In (b) phase compensation system operates to restore 99.6 % of power to the carrier. Magnification by 15,000 in (c) shows linewidth after correction is below 1 mHz!

able to transfer the stable frequency reference needed for our frequency synthesis project.

Recently we reported[13] a new form of lineshape distortion with FM spectroscopy of Rb atoms using a frequency-stabilized Ti:Sapphire laser. The effect is easily observable for a range of modulation frequencies from 1-10 units of the linewidth, and is manifest as a phase-shift of the received rf modulation on the probe beam with increasing probe-beam intensity. The phase-shift arises when the atomic dipole vector begins to see the phase modulation of the carrier as important relative to the natural relaxation rate. Theoretical calulations based in the carrier-frequency rotating frame led to a clear understanding of the intensity-dependent phase as a phase lag of the dipole response to the carrier's phase perturbation associated with the modulation process. These effects of non-equilibrium response are very general and should be present in all spectroscopic methods with time-modulated fields. Superposed on them are the changes associated with momentum transfer to the system, which can be controlled by working with an open system (I_2) or a closed system ("cycling" transitions in alkali metals' D_2 lines.) A full theoretical framework for understanding these combined effects has been developed by our consultant and long-time colleague, Prof. Christian J. Bordé of the Laboratoire des Physique des Lasers, Paris. Via lineshape analysis and use of transitions differing in their degree of "openness," we expect to be able to isolate these physical processes for study.

The research on active control of vertical acceleration and tilt of the optical table in the "Quiet Room" has made major progress during this period, due substantaially to the use of the modelling program "Matlab" by an experienced former student. We are using electronic bubble tiltmeters for dc tilt information, a sensitive seismometer for vertical pickup, and 3 proximity sensors for fast floor reference and dc height stabilization. The actuators for the servo are time-proportioning valves to control the air legs, and small electromagnetic thrusters to provide vertical control beyond 150 Hz. Some further refinement is needed to orthogonalize these channels, but this setup is nearly working at the desired levels (< 1 nano-"g" vertical, and < 0.1 μrad tilts). Unfortunately for us, the young researcher was offered a position with one of the major manufacturers of optical tables to continue this work under more attractive financial conditions. To bring the system at JILA to a full operational level, we need to obtain a fast computer with DSP capability to implement the control loop algorithm in a software-based environment to facilitate rapid and well-characterized changes.

For completeness we briefly summarize a number of other advances and activity areas:

1) Continuing development of a technique and apparatus for accurately measuring the fractional fringe order of Fabry-Perot ring fringes. Our scientific application will be the precise measurement of Fabry-Perot rings to obtain accurate laser

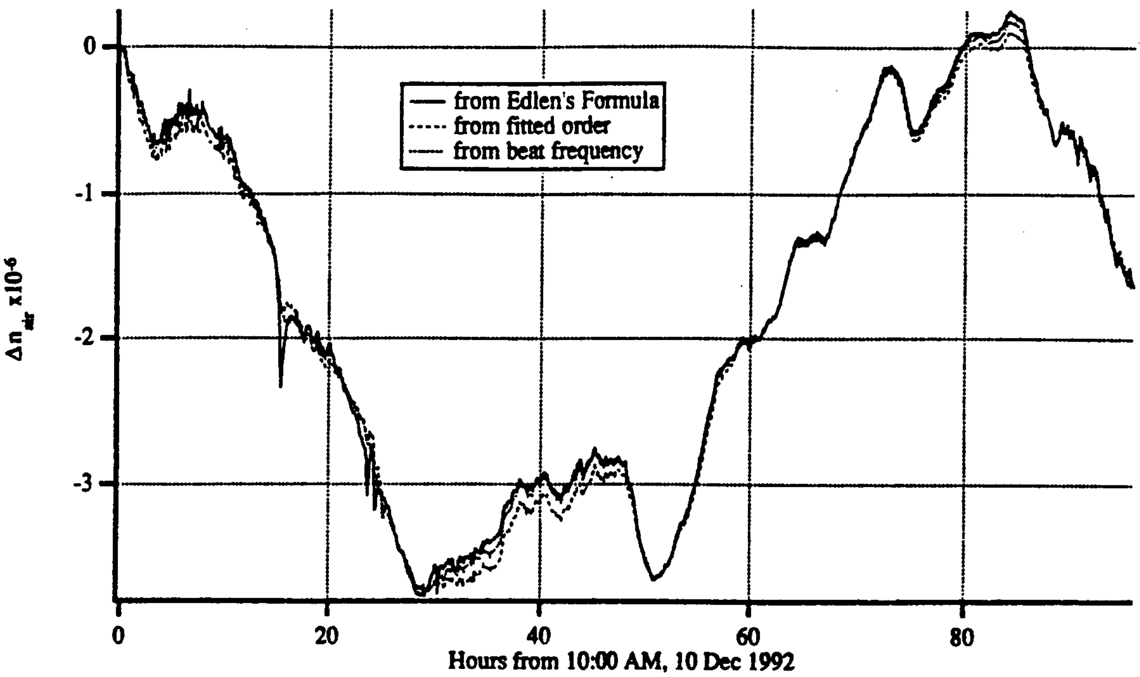

Figure 5. Comparison of our two interferometric methods with air index calculated from Edlén's formula.

wavelengths when tuned to some interesting atomic transition. An important engineering application of this interference-phase measuring capability is the readout of atmospheric refractivity for industrial interferometric length/position control applications. Our software gives 2×10^{-4} rms orders *accuracy* in fringe fitting of synthetic data constructed with 5% rms random noise. Experimental confirmation has been shown to 1×10^{-3}, limited by small systematic deviations of the actual fringe lineshapes relative to the expected Airy fringes, likely caused by residual wedge and other optical defects in our interferometer. An extended comparison of three approaches to real-time determination of atmospheric refractivity has been carried out, with agreement at the 10^{-7} level, and some systematic deviations which may point to humidity retention in/on one of the coatings. This work formed part of the basis for the thesis of M. L. Eickhoff and a paper is almost ready for submission. The refractometer system patent was filed by NIST on July 1, 1991 and issued June 8, 1993 as US Patent # 5,218,426. NIST is now seeking licensees for application of the refractometer, for example as an adjunct measurement tool for on-line use in semiconductor wafer fabrication and other manufacturing facilities where high accuracy distance measurements are made by interferometry in the ambient air.

2) Improved phase stabilization of low-cost laser diodes using external resonators and fast electrical feedback. A nice method for measuring the diode's current/frequency transfer function was developed, and is being written up for publication in RSI. Optimal equalizers were designed to allow substantial increase of the servo bandwidth to ~10 MHz, which allowed capture of ~99.8 % of the emitted power in the phase-locked spectral delta-function. This work was done in collaboration with a former JILA student, Kurt Gibble, during his postdoc time at Stanford, and with Robert Wynands, a student in the Max Planck group of Professor T. W. Hänsch in Munich.

3) Efficient doubling and Optical Parametric Oscillator based on KTP and $KNbO_3$. This efficient doubling research began at JILA, but this demonstration was carried out at Cal Tech, beginning as part of our collaborative project, but finally pursued to a very successful conclusion by Eugene Polzik, Jeff Ou, and Jeff Kimble, using some JILA-made electronics. For new quantum optics experiments at JILA we are exploring the use of type II-matching in KTP to form an OPO pumped by a 514 nm Argon laser. We have seen parametric fluorescence but no parametric oscillation has been obtained so far.

4) Promising results have been obtained on a 612 nm HeNe system, stabilized to an external cavity containing an Iodine cell. The apparatus is expected to provide frequency stability and reproducibility in the 10 Hz level in the size of an airlines-carry-on package. It is a joint project with the BIPM in Sévres, France. This project is temporarily on hold pending completion of the absolute frequency measurements in the green.

4 b. Special results. Probably the optical frequency measurement of the stabilized Nd laser is the most dramatic result of the recent work. We are excited about the prospects of these various pieces individually, but taken together it is clear that remarkable progress lies just ahead!

5. Extenuating Circumstances. This JILA work has long been jointly supported by NIST, NSF, and the ONR, with occasional bursts of support from AFOSR. Rather large fluctions of the input are thus expected and observed. On the consumption side, with four large-frame argon lasers employed in these experiments, we experienced a –55 K$ transient last year for necessary replacement of two plasma tubes. One more is just at the edge of death. A reasonable way to address such fiscal fluctuations in the short term is to define some collaborative research programs with other institutions. This approach includes colleagues at the Institute for Laser Science, Tokyo, in a collaboration about laser-trapped rare gases and alkaline earths. We expect to continue our collaboration also with Professor Teo Hänsch in Munich relative to the frequency measurement scheme to compare microwave and optical frequencies, the so-called "Divide and Conquer" system. This latter channel, supported by the Humboldt Stiftung, is an attractive way to make use of another organization's facilities for high-risk experiments. Quantum optics continues to be pursued mainly as a collaboration with Professor H. J.

Kimble at CalTech. One can project another 4 or 6 week interaction on ultrastable lasers with the Virgo gravitational-wave detector collaboration in Orsay and Pisa. While in a limited way these external activities may make economic sense, in the long run this miserly approach toward risk acceptance has a depressing effect on the people involved in our more programatic research. Either drastic scaling back or additional funds or both will be necessary to restore optimal operation in Boulder.

6. Other Government Support: Other funding support for this work is supplied by the National Science Foundation as a portion of the block grant to JILA for research in atomic, molecular and optical physics. The portion available for this work varies considerably and this time was sufficient to support three student salaries and some shop work; approximate total amount $150 K. Ordinarily a principal source of support is the National Institute for Standards and Technology which normally supplies about $50 K for continuing research on laser stabilization and precision measurements. We also had a $170K pa two-year funding from AFOSR, Nov 1, 1991-Oct 30, 1993, mainly supporting the work of L. Hollberg. Another AFOSR grant for $ 100 K pa, to L. Hollberg and the present PI, is pending.

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7. Publications 1991-1994:

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3. J. L. Hall, "Frequency stabilized lasers -- from the beginning toward the future," invited submission to Laser Physics (Moscow) (Jan 1994) for memorial issue honoring V. P. Chebotayev.

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5. M. P. Winters and J. L. Hall, "The Correlated Emission Laser - An Experimental Investigation," accepted by Physical Review A.

6. M. Zhu and J. L. Hall, "Short and long-term stability of optical oscillators," subm. to IEEE Trans. on Ferroelectrics, Ultrasonics, and Frequency Control.

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1. J. L. Hall, M. Zhu, and D. Hils, "Dreams about the next generation of super-stable lasers," in *Laser Spectroscopy 10,* (E. Giacobino, G. Camy, and M. Ducloy, Eds., World Sci. Publ., 1992) .

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3. M. Zhu, C. W. Oates and J. L. Hall, "White light cooling of an Atomic Beam," Conference on Quantum Electronics Laser Science, Baltimore MD, May 12-17, 1991, Digest p. 118-9.

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Patent Disclosures

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